

Voltage-induced broad-spectrum reflectivity change with surface-plasmon waves

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Voltage-induced broad-spectrum reflectivity change with surface-plasmon waves is reported. When white light is incident at a metal/electro-optical material interface, surface-plasmon waves can be excited under phase match conditions. This surface-plasmon resonance depends on the dielectric constants of both the metal and the electro-optical material. Photons in the surface-plasmon resonance wavelength range are absorbed by the interface. Since metals have large imaginary parts of their dielectric constants, the surface-plasmon resonances are broad and may cover all visible wavelengths. Applying voltage to the electro-optical material to change its dielectric constant can result in a change in the reflectivity at the interface. Experimental results showed a reflectivity change from almost 0% to about 40% under an applied voltage using a liquid-crystal and nickel film structure, and the results had good agreement with theoretical calculations. The theoretical calculations also predicted a 90% reflectivity recovery by exciting surface-plasmon waves at a Rh–Al/electro-optical material interface. These results demonstrate that a high efficiency white light modulator can be built using surface-plasmon excitations. © 2005 American Institute of Physics. [DOI: 10.1063/1.1830093]

I. INTRODUCTION

Surface-plasmon (SP) waves are transverse magnetic (TM) electromagnetic waves which propagate along the interface of a metal and a dielectric. SP waves propagate parallel to the interface and are exponentially attenuated in the normal direction in both the metal and the dielectric. Optical excitation of SP waves was demonstrated by Otto¹ using a frustrated total reflection. Kretschmann² showed that SP waves could be excited in a thin metal film evaporated onto the base of a totally reflecting quartz half cylinder. Since then, the attenuated total internal reflection (ATR) has been widely used to investigate the nonradiative SP waves on metal surfaces. SP waves can be described as a collective oscillation in electron density at the interface of metal and dielectric.³ At SP resonance, the reflected light attenuates. SP resonance is dependent upon the dielectric constant of both the metal and the dielectric. By changing the refractive index of the dielectric, we can change the SP coupling efficiency and resonance frequency.

When SP waves are excited at a metal/electro-optical (EO) material interface, reflectivity at the interface can be changed by applying a voltage across the EO material. Many studies have involved modulation of the ATR spectrum associated with the excitation of SP waves. This effect was demonstrated by using a piezoelectric transducer to mechanically modulate the air gap in the Otto configuration for SP excitation.⁴ The Pockels response of a monolayer Langmuir–Blodgett film was used to produce SP modulation in a Kretschmann configuration.⁵ Wang and Simon⁶ reported that

when an ATR geometry was used in which a quartz electro-optic crystal onto which a thin Ag film had been evaporated was in contact with the hypotenuse face of a high-index prism, modulation in the ATR of a He–Ne beam near the angle of SP excitation was observed when a low-frequency electric field was applied to the crystal. Experiments have been done with single and multiple boundary SP waves; the angular spectra show phase reversal structure due to the interference of the modulated reflected wave with the ATR wave. Yeatman and Caldwell⁷ reported a SP spatial light modulator based on nematic liquid crystals. Their work indicated that the spatial resolution of a SP light modulator is related to the plasmon decay length, which is about 10–20 μm at an incident wavelength of 633 nm for silver. They achieved a contrast ratio greater than 100:1 and claimed that for a SP liquid-crystal light modulator, its switching time could be 50 times faster than the bulk switching time of the liquid-crystal cells. The application of the long-range SP mode for electro-optic modulation have also been proposed.⁸

Most of the SP light modulator studies were focused on monochromatic light modulation. Considering a SP light modulator with incident white light, the photons in the SP resonance wavelength range are absorbed and the photons out of the resonance wavelength range are reflected. When a voltage is applied to the EO material, the dielectric constant of the EO material changes, and the SP resonance wavelength also changes. Therefore, an electronic wavelength selection can be achieved by shifting the SP resonance with the applied voltage.

In 1995, Wang reported that SP waves could generate voltage-induced color-selective absorption.⁹ When white

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light was incident at a silver/liquid-crystal interface, those photons in the SP resonance frequency range were absorbed, and the reflected light showed the complementary color. In that report, when an applied voltage across the liquid crystal increased from 0 to 30 V, the SP absorption spectrum shifted from 650 to 450 nm. recently, Wang reported that the coupled SP waves could also generate wavelength selection,¹⁰ and this wavelength selection would depend on the coupling distance of the two SP waves. Experiments showed that when the coupling distance changed from 50 to 1000 nm, the peak transmission shifted from 414 to 852 nm without harmonic peaks. Theoretical modeling showed that the tunable range of the peak transmission could be extended from 400 to 1600 nm.

It is well known that the SP wave-vector dispersion at a metal/dielectric interface can be written as

$$k_x = \frac{\omega}{c} \sqrt{\frac{\epsilon_1 \epsilon_2}{\epsilon_1 + \epsilon_2}}. \quad (1)$$

Here, k_x is the tangential component of the wave vector, ϵ_1 is the dielectric constant of the metal, ϵ_2 is the dielectric constant of the dielectric, ω is the frequency of the incident light, and c is the speed of light. For most metals ϵ_1 is less than -1 in the visible region of the spectrum, and we see that k_x is greater than the wave vector of an electromagnetic wave in the dielectric at the same ω . SP waves may be excited only with evanescent waves and therefore classified as nonradiative. In order to generate SP waves optically, they must be coupled to radiative modes. This can be accomplished using prism coupling with geometry proposed by Kretschmann.

For prism coupling, Eq. (1) can be written as

$$n \times \sin \theta = \sqrt{\frac{\epsilon_1 \epsilon_2}{\epsilon_1 + \epsilon_2}}. \quad (2)$$

Here, n is the index of the prism and θ is the incident angle at the prism/metal interface. Many metals exhibit a very large dispersion in the optical frequency, while the dispersion of most of the dielectric materials is relatively small in the same range. If a liquid-crystal material is used as the dielectric, it can be assumed to the first-order approximation that the dielectric constant of the liquid-crystal ϵ_2 is only a function of an applied voltage V , i.e., $\epsilon_2 = \epsilon_2(V)$. Likewise, the dielectric constant of the metal is only the function of the optical frequency ω , i.e., $\epsilon_1 = \epsilon_1(\omega)$, and the index of the prism is a constant. Equation (2) can be rewritten as

$$\frac{1}{\epsilon_1} = -\frac{1}{\epsilon_2} + \frac{1}{n^2 \sin^2 \theta}. \quad (3)$$

Many metal materials exhibit a very large dispersion in the optical frequency range, while the dispersion of most of the dielectric materials is relatively small in the same range. For the first-order approximation, we can assume that the dielectric constant of the liquid crystal is independent of the optical frequency and only a function of the applied voltage. Then the voltage-induced SP resonance frequency change can be derived as

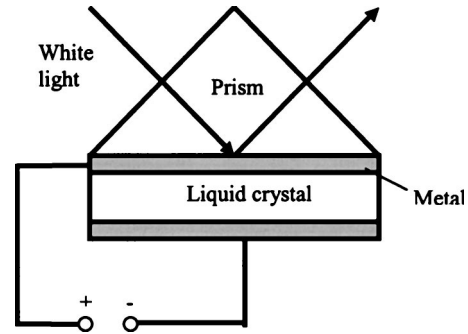


FIG. 1. Experimental setup used to measure the voltage-induced reflectivity change with surface-plasmon waves.

$$\frac{d\omega}{dV} = \left(\frac{\epsilon_1}{\epsilon_2} \right)^2 \frac{d\epsilon_2}{dV} \left(\frac{d\epsilon_1}{d\omega} \right)^{-1}. \quad (4)$$

This relation indicates that the voltage-induced surface-plasmon resonance frequency change is dependent upon the dielectric constants of both the metal film and the liquid-crystal material, the voltage-induced dielectric constant change, and the dispersion of the metal film. From Eq. (4), we can see that the voltage-induced frequency change is proportional to the inverse of $(d\epsilon_1/d\omega)$. In other words, if the metal has a large change over a certain optical frequency range, then we can have a large voltage-induced SP resonance change, for example, from visible to infrared (IR), with a relatively small voltage added. A more detailed calculation can be derived by a linear reflectance from multilayer in the ATR regime. The reflectance of the prism-metal-dielectric system may be written as

$$R_{012} = \frac{r_{01} + r_{12} \exp(i2k_{1z}d_1)}{1 + r_{01}r_{12} \exp(i2k_{1z}d_1)}. \quad (5)$$

Here r_{ij} are the Fresnel reflection coefficients for p -polarized light and k_{1z} is the normal component of the wave vector. The r_{ij} are given by

$$r_{ij} = \frac{\sqrt{\epsilon_j} \cos \theta_i - \sqrt{\epsilon_i} \cos \theta_j}{\sqrt{\epsilon_j} \cos \theta_i + \sqrt{\epsilon_i} \cos \theta_j}, \quad (6)$$

where the subscripts $i, j=0, 1, 2$ refer to the prism, the metal, and the liquid crystal, respectively, and d_1 is the thickness of the metal film.

Here, we report an experimental result of the voltage-induced broad-spectrum reflectivity control with SP waves. When the white light is incident at a Ni/liquid crystal interface, and a voltage is applied across the liquid crystal from 0 to 60 V, the reflectivity increases from 0% to near 40%. Theoretical calculations have shown that if we use a Rh-Al two layer film to replace the Ni film, the reflectivity could change from 0% to near 90%.

II. EXPERIMENT

The samples were prepared by thermally evaporating aluminum and nickel films onto the hypotenuse faces of two 60° SF57 glass prisms, respectively. A 50-nm MgF₂ layer was then evaporated at a 50° oblique angle off the normal onto the metal films as the liquid-crystal alignment layer. The

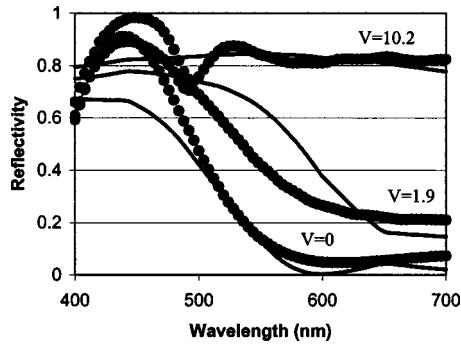


FIG. 2. Voltage-induced reflectivity change with a 19-nm Al film (dotted, experimental; solid, theory).

same alignment layer was evaporated onto an indium-tin-oxide (ITO) glass plate as the other electrode. A cell was assembled with alignment directions parallel, separated by 4- μm glass spacers, and filled by a capillary action with a nematic liquid crystal (Merck BL009).

The experimental setup, using the Kretschmann geometry, is shown in Fig. 1. A p -polarized collimated white light beam was incident on the metal/liquid-crystal interface through the coupling prism at an angle beyond the critical angle, a dc voltage was added across the liquid crystal, and the reflected light was projected onto a white screen and measured by a Spectrascan PR650 made by Photo Research.

The first experiment was performed with a 919-nm aluminum film. Its spectral reflectivity change as a function of the applied voltage is shown in Fig. 2. Here the incident angle of the prism/Al interface was 61° . The dots are experimental data and the solid curves are theoretical calculations. Without the applied voltage, the SP resonance covered all of the green and red color spectral ranges, as almost all of the red and green photons were absorbed. Under this condition the blue photons exhibit reflection near 90% at 440 nm with a corresponding chromaticity of the reflected light appearing deep blue. When the applied voltage was increased to 1.9 V, the liquid crystal had an effective index change of $dn=0.01$, and the SP resonance shifted toward the infrared (IR). The reflected light exhibited a light blue chromaticity with about 20% reflectivity for green and red photons. When the applied voltage was increased to 10.2 V, the liquid crystal had an effective index change of $dn=0.04$, and the SP resonance

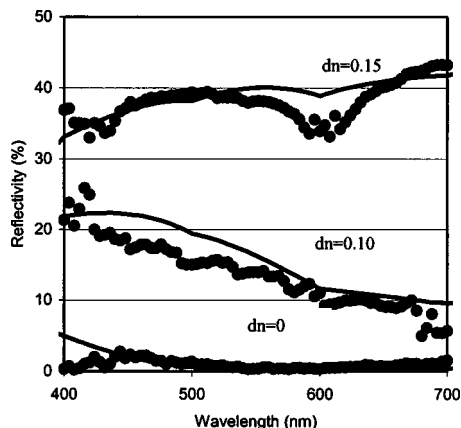


FIG. 3. Voltage-induced reflectivity change with a 22-nm Ni film (dotted, experimental; solid, theory).

TABLE I. Optical properties of aluminum.

Energy (eV)	1.50	2.00	2.40	3.00
Wavelength (nm)	826	620	517	413
n	2.75	1.30	0.628	0.523
k	8.31	7.48	6.24	5.02

shifted further toward the IR. The reflected light was a broadband with the appearance of bright white. The experimental data (dots) agree with the theoretical calculations (solid lines) very well.

The second experiment was performed with a 22-nm-thick nickel film. Its spectral reflectivity as a function of the applied voltage is shown in Fig. 3, where the dots are experimental data and the solid curves are theoretical calculations. Here the incident angle was set at 70° . When p -polarized white light was incident at the glass/Ni interface at zero voltage, the SP resonance covered the entire visible spectrum and almost no visible light was reflected. When 30 V was applied across the liquid crystal, the liquid crystal had an index change $dn=0.01$, and the SP resonance shifted to the IR, resulting in a reflectivity of about 20%. When the voltage was increased to 60 V, the liquid crystal had an index change $dn=0.15$, and the SP resonance shifted further away from the visible, and the reflectivity reached near 40%. Once again, the experimental results were in good agreement with the theoretical calculations.

The refractive indices of the liquid-crystal BL009 used in the calculations are from the Merk liquid-crystal catalog. The optical constants of aluminum and nickel used in the calculations are listed in Tables I and II, respectively.¹¹

III. RESULTS AND DISCUSSION

When a SP resonance is excited, the photons in the SP resonance range are removed from the reflected spectrum by resonant absorption. The SP resonance bandwidth strongly depends on the dielectric constant of the metal film. The dielectric constants of metals have real parts and imaginary parts. Generally speaking, the real part of dielectric constant of a metal determines the SP resonance angle, and the dispersion of the real part and the value of the imaginary part of dielectric constant of a metal determine the bandwidth of the SP resonance. For metals with large dispersion of the real part of and small value of the imaginary parts of their dielectric constants, their SP resonances are relatively narrow. For example, the experimental result of single SP resonance with silver film shown in Ref. 9 had a full width half maximum (FWHM) about 50 nm, and the experimental result of coupled SP resonance with silver shown in Ref. 10 had FWHM about 30 nm in the visible spectrum. For metals with

TABLE II. Optical properties of nickel.

Energy (eV)	1.50	2.00	2.50	3.00
Wavelength (nm)	826	620	496	413
n	2.53	1.92	1.67	1.61
k	4.47	3.65	2.93	2.44

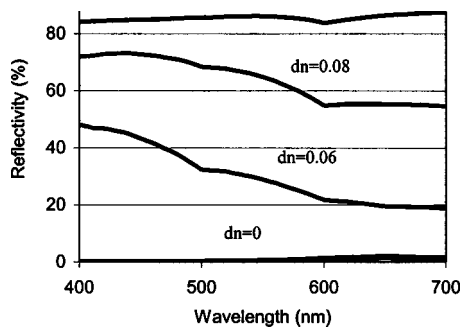


FIG. 4. Theoretical calculations of voltage-induced reflectivity change with Rh-Al two layer film.

larger imaginary parts of their dielectric constants, for example Al and Au, the SP resonance spectrum is broader. For metals with very large imaginary parts of their dielectric constants, for example, Rh, Ni, or Pt, the SP resonance can become so wide that it may cover the entire visible spectrum. When a voltage is applied upon the EO material, the SP resonance can be shifted from the visible to the IR, and the absorption of the visible photons can be reduced, and the reflected light can resume. This variation in white light reflectivity can then be employed as a white light modulator or an electronically tunable mirror.

Twisted nematic liquid-crystal materials have a positive dielectric anisotropy, when a voltage is applied, the director of their molecules in the center of the layer tends to orient itself parallel to the applied voltage. When the applied voltage is above some threshold, the molecules of the entire layer, except for those bound at the immediate surface of the alignment layer, become oriented with the directors everywhere parallel to the applied field. But SP wave is a surface wave, the surface layer of the liquid crystal, which is bounded by the alignment layer, and has the most contribution to the SP resonance. Therefore, the effective refractive index change is not a linear function for our SP white light modulator, and the uniformity of the liquid-crystal alignment can generate structures in these experimental curves.

Theoretical calculations showed that larger reflectivity recovery could be achieved with a rhodium (Rh) film. Theoretical calculations also showed a Rh-Al two layer film can have a reflectivity change from almost 0% to near 90%, as shown in Fig. 4. When the voltage-induced index change of the liquid-crystal $dn=0$, the SP resonance covers all of the visible spectrum, and the reflectivity is near zero. When a voltage is applied to induce an index change of the liquid crystal to $dn=0.06$, the SP resonance shifts toward the IR, and the reflectivity increases to about 40% in the blue and 20% in the red; when the index change of the liquid crystal is $dn=0.08$, the reflectivity increases to about 70% in the blue and 50% in the red; and when the index change of the liquid crystal reaches $dn=0.10$, the reflectivity increases to near 90% over the visible spectrum. The Rh layer provides a SP resonance frequency over a broad range, covering the entire visible spectrum, and the Al layer enhances the maximum reflectivity when the SP resonance is shifted out of the visible spectrum. The optical constants of rhodium used in the calculations are listed in Table III.¹¹

Though the liquid-crystal material was used in this ex-

TABLE III. Optical properties of rhodium.

Energy (eV)	1.50	2.00	2.50	3.00
Wavelength (nm)	826	620	496	413
n	2.78	2.12	1.88	1.53
k	6.67	5.51	4.65	4.29

periment, practical device structures could also employ solid-state EO materials, such as lead-lanthanum-zirconate-titanate (PLZT), potassium dihydrogen phosphate (KDP), potassium titanyl phosphate (KTP), EO polymers, aurivillius oxides, organic crystals, and organic salts. The potential advantages of a solid-state material include high-speed, voltage-induced reflectivity change (expected to be three orders of magnitude faster than the nematic liquid crystals), compatibility with thin-film processing techniques, and improved device packing.

IV. CONCLUSION

In conclusion, voltage-induced broad-spectrum reflectivity change with SP waves was demonstrated, with good agreement between experimental and theoretical calculations. For metals having a large imaginary part of their dielectric constant, the SP resonance spectrum can be sufficiently wide such that it can cover the entire visible spectrum. Experiment showed that when a SP resonance was generated at a nickel/liquid-crystal interface, a white light modulator can be built. When a voltage is applied to change the index of the liquid crystal, the reflectivity can change from almost 0 to up to about 40% over the entire visible spectrum. Theoretical calculation also predicted a 90% reflectivity recovery by exciting surface-plasmon waves at Rh-Al/electro-optical material interface. A high efficiency white light modulator can be built upon the SP light modulations discussed above.

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